Properties of sediments deposited in a fluid with different pH

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Abstract

The effect of pH on the physical and mechanical properties of a sediment was investigated through a set of experimental tests. The sediment was formed from deposition of suspended particles in a fluid. Two different types of clay soil were suspended in fluids with different pH (2, 4, 7, 9 and 11) in cylindrical tubes with volume of 1 liter and also in special cylindrical reservoirs. The height of the sediment was measured in the cylindrical tube until equilibrium was achieved. The sediment deposited in the reservoirs was dried in air and then Atterberg limit, compaction and unconfined compressive strength (UCS) tests were conducted on samples prepared from each sediment. The results showed that the final height of the settled sediment is a function of pH; the height of sediment is increased with increasing the pH. Also, the Atterberg limits increased with increasing the pH. The maximum dry unit weight and optimum water content decreased and increased with increasing the pH. The final strength of the sediment decreased with increasing pH. Based on SEM analysis, it was found that the values of pH influence the properties of the formed sediments.

Key words: Sediment, pH, Atterberg limits, Compaction, Strength

Introduction

Soils are originated by mechanical, physical and chemical weathering of rocks. The type of weathering has a very different effect on productive soils. The productive soils can either remain in situ or be transported elsewhere by water, glaciers, wind or gravity to form deposits. Water is more important than the other transporting agents and is responsible for most of sedimentary deposits. Soil particles can be carried by water when the velocity of water is high. When streams or rivers with suspended particles enter a lake or sea, the sediment load is deposited in grading order. The deposited soil can be divided into delta deposits, estuarine deposits and marine deposits. These sediments are different in particle size and behavior. The marine soils are deposited on the continental shelf and are finer in size.

Sedimentation or deposition is the tendency of soil particles to settle out of suspension and is a function of particle size, shape, density, and fluid density and viscosity (Van Olphen, 1964, Benna et al., 1999 and Bessho and Degueldre, 2009). Sridharan and Prakash (1999) reported that the clay mineralogy and pore water chemistry influence the behavior of deposits. They stated that various forces may be involved in the settling of suspended particles of which, they are subjected to three main forces. These are the force due to self weight, and electrical forces due to attraction or repulsion. In coarse grained soils, the electrical forces are negligible compared to the forces due to the self-weight, but they play an important role in controlling the settling behavior of fine particles. Sridharan and Prakash (2001) stated that, when there is no mutual interaction between the settling particles, the dispersed free settling results in segregated sediments which are

characterized by the existence of coarse particles at the bottom of sediment and the fine particles at the top. However, the type of clay particles that exist in the soil can play an important role during the settlement process. The mechanical and physical properties of these deposited soils are influenced by the environmental conditions. Imai (1980) stated that the degree of flocculation and mutual interaction between soil particles control the settling form of deposits. He also indicated that the settling behavior of suspended clay particles is a function of initial water content and ionic concentration. Ohstubo and Ibaraki (1991) showed that the electrical charges of suspended particles is a factor that controls the settling characteristics of a soil. Palomino and Santamarina (2005) presented a fabric map for kaolinite as a function of salinity and solution pH, to explain the factors that influence particles flocculation and sedimentation. They indicated that sedimentation at low salt concentration is a function of pH. Kaya et al. (2006) showed that flocculated and dispersed settling are two extreme characteristics for soil particles and depend on the clay mineralogy, ionic strength and solid to water ratio that control the settling process of soils in aqueous environment. They also showed that the zeta potential can be another factor in settling of suspended particles. Chen and Anandaraj (1998) and Palomino and Santamarina (2005) stated that the relative change in magnitude between the attractive and repulsive forces due to Van der Waals forces causes the initiation of flocculated particles and ultimately sedimentation, as flocculated clay particles are heavier and settle faster relative to individual particles.

Clay soils have two structures, namely micro-structure and macro-structure. The physical and chemical interactions that are influenced by active clay minerals occur within microstructure. Significant information about the structure of soils can be obtained with

the advent of scanning electron microscopy (SEM). SEM is now widely used for explanation of different types of soil behavior (Collins and McGown 1974).Tremblay et al. (2002), Botta et al. (2004), Cai et al. (2006) and Estabragh et al. (2016) used SEM analysis for observing the microstructure of soil samples under different conditions and for explaining their observed behaviors.

Extensive amount of research has been performed on physical and mechanical properties of deposited sediments in river basins, lakes and seas (e.g., Wang et al., 2005, Sheahan and De-Greet, 2008 and Liu et al. 2017). In arid and semiarid regions of the world, there are usually seasonal rivers and lakes where the sediments deposited in their bed are dried due to evaporation in the summer. On the other hand, sometimes to facilitate shipping, the sediments of a river are dredged and dried in air. Sediment deposition in harbors is also a problem that is sometimes faced by marine engineers as it affects the usefulness of harbor. Again, dredging may be a feasible solution to these problems. These dredged sediments are sometimes used for construction of roads, embankments, buildings, etc. In general, such deposits consist of fine grained soils such as clay and silt and their characteristics are very important when they are used as construction materials. Therefore, it is important to be able to predict the behavior of these sediments in order to guarantee the safety of projects constructed with or on them. In this work, the physical and mechanical properties of two clay soils that were deposited in an aqueous fluid with different pH values are studied through a program of experimental tests. In what follows the results of the experiments on the sediments are presented and discussed.

Materials and experimental methods

The main materials that were used in this program are soil and fluid with different pH values. The properties of these materials are described in this section.

Soil

Two types of clay soil were used in this work that are hereafter referred to as soil A and soil B. The physical and mechanical properties of these soils were determined according to ASTM standard and are shown in Table 1. According to the Unified Soil Classification System (USCS), both soils (A and B) can be classified as clay with high plasticity (CH).

Water

Drinking water was used in this work. It had a pH of 7.0, chloride content of 17 meq/L and calcium+magnesium content of 9.1 meq/L. By adding a predefined volume of HCl or NaOH to a volume of drinking water, solutions with desired pH values of 2, 4, 9 and 11 were prepared for the sedimentation process. In other words, the acidic and basic aqueous media were prepared by controlling the pH.

Sediment and sample preparation

The soil samples used in this study were natural soils (soil A and B). Soil sediment depositions were conducted in water with pH of 7 and in solutions with pH of 2, 4, 9 and 11. The sedimentation process was carried out in a one-liter sedimentation tube and a circular reservoir with 1.6 m diameter and 0.4 m height. It should be noted that the sizes of the sedimentation tube and reservoir have no effect on the settling characteristics of soil as indicated by Michaels and Bolger (1962).

Selection of the ratio of solid to water is an important factor in controlling the settling process of soils in aqueous environment (Imai, 1980 and Kaya et al., 2006). The ratio of solid was chosen as 5% weight of the mixture (fluid and solid) that is similar to the

recommended value for hydrometer testing (ASTM D 422-63) and used by Gorakhki and Bareither (2015).

The required quantity of air dried soil was added to the reservoir in increments and mixed thoroughly to reach a dilute soil-water mixture. For the other test series, HCl and NaOH were added to water and mixed thoroughly to reach the solution with the desired value of pH (2, 4, 9 and 11). The soil was then added to each of these solutions as in the previous stage and mixed to obtain a dilute mixture of soil and solution water. This dilute mixture was then allowed to settle for more than two weeks. After that the excess liquid above the soil was drained off, the soil was then air dried and the desired samples were prepared from this sediment for the experimental tests. Standard compaction tests, according to ASTM D 698-07e, were conducted on the sediments deposited in normal water and in different solutions. The maximum dry unit weight and optimum water content were determined for each soil. The samples for unconfined compressive strength test were prepared from the different dry sediments at the optimum water content and maximum dry unit weight obtained from the compaction test. For preparing the soil samples, the grounded sediment was mixed with the required amount of water corresponding to the optimum water content. Static compaction method was used for preparing the samples. Compaction was done in a special mould by using a compression loading machine. Compaction was conducted in three layers of grounded sediment with optimum water content until that the maximum dry unit weight corresponding to compaction curve for each sediment was achieved. The value of the applied load on each layer was set to the amount that was needed to create the maximum dry unit weight of desired sediment. This vertical load was determined by trial and error and was used in compaction. Each layer

was compacted to the pre-defined vertical load at a fixed displacement of 1.5 mm/min until the maximum dry unit weight was achieved. The prepared samples were 50 mm in diameter and 100 mm long.

Test Program

Suspension tests were conducted on samples of natural soil (soils A and B). In these tests 50 g of natural dry soil was mixed with 125 cm^3 liquid with desired pH (i.e. 2, 4, 7, 9 or 11) and transferred in an electric mixture for duration of about 30 minutes. The mixed solution was poured in a cylinder and then more desired fluid was added to bring the total volume of the mix to 1000 cm^3 similar to the method that was used by Sridharan and Parakash (2001) and Kaya et al. (2006). The top of the cylinder was then covered and it was shaken by hand for a few minutes in order to ensure that the soil particles were completely mixed in the solution. The soil was then allowed to settle until it reached the condition that no changes in the sediment thickness were observed. During the settlement, the height of the sediment was measured with accuracy of 1.0 mm. Atterberg limits, compaction and unconfined compression tests were conducted on the samples of sediments (that were deposited in different fluids with various pH values) according to the ASTM standard. For unconfined compression tests the samples were loaded in a compression loading frame at an axial displacement rate of 1 mm/min. The applied load was recorded continually and the tests were terminated when the failure of samples was attained.

Scanning electron microscopy (SEM) tests were performed on the samples in order to observe the microstructure of the samples in different conditions. The samples were prepared according to the optimum water content and maximum dry unit weight. After

that the samples were carefully cut with a sharp knife into small pieces of disk shape with 1 cm^3 volume (as used by Tremblay et al. 2002 and Estabragh et al. 2016) and scanned under SEM.

Results

The sedimentation of soils A and B with time in different solutions is illustrated in Figs 1-a and 1-b respectively. As shown in these figures, the initial height of the suspended particles for soils A and B is about 340 mm. The thickness of sediment decreased with time because of the deposited suspended particles until it reached to equilibrium condition. The results show that the settlement of the suspended particles for each soil is dependent on the value of pH. When the pH was basic (9 or 11) the settlement was slower than the cases when the solution was acidic. For example, for soil A when the pH was 11, the settlement of suspended particles occurred after 2.16 hours but at pH of 2, it occurred after 0.533 hours. The results show that the trend of settlement for soil B was similar to soil A, but the beginning of settlement at a particular pH was not the same. As it is seen from Fig. 1b, the starting of settlement at pH of 11 is about 0.533 hours after the start of the test. Therefore, it is seen that the settlement of suspended particles is a function of pH for both soils but the starting time of settlement is dependent on the type of soil.

Fig.2 shows the Atterberg limits including liquid limit (LL) and plastic limit (PL) for the sediments resulting from settlement of suspended particles of soils A and B in acidic and basic solutions. As shown in this figure, the values of both LL and PL are increased with increasing the pH of the fluid. The LL and Pl for soil A at pH of 2 are 64.2 and 23 % but at pH of 11 they change to 91 and 42% respectively. This trend of variation is also

observed for soil B but the rate of these variations is not the same as soil A. At pH of 2, the LL and Pl for soil B are 64 and 27% but at pH of 11 they change to 72 and 34% respectively. The difference in the rates of variation of LL and Pl for the two soils may be due to the initial composition of the formed minerals for each soil.

Figs.3a and 3b show the compaction curves for the two soils at different values of pH. As shown in this figure, for both soils in acidic conditions, the compaction curves moved to the right hand side of the compaction curve for the solution with pH of 7, but in the basic condition, it moved to the left of the compaction curve for pH of 7. These results indicate that the compaction curves for sediments are a function of pH of the aqueous medium. For the sediments that were deposited in basic solution, both the maximum dry unit weight and optimum water content decreased in comparison with the fluid at pH of 7 while the opposite trend was observed for the acidic solutions. It can be concluded from comparing the results of the compaction tests for soils A and B that at a given value of pH, the compaction parameters (maximum dry unit weight and optimum water content) are not the same for the two soils. This could be due to the difference between the two types of soil.

The stress-strain curves for the deposited sediments at various pH values are shown in Figs. 4a and 4b for soils A and B. As shown in these figures, the stress-strain curves evolved by decreasing the value of pH. In other words, in acidic aqueous condition, the strength of sediment was increased but in basic condition the strength was decreased. For soil B the final strength at pH of 2 was 306 kPa but when pH was 11, it was changed to 257 kPa. This shows a reduction of nearly 17%. Similar trends of variations in strength with pH was observed for soil B. The results show that the failure stress of soil A

occurred at strain of 6% but for soil B it was at strain of 3%. This indicates that the ductility of soil A is more than soil B.

Figs 5 and 6 show the micrographs for the deposited sediments at different pH values for soils A and B respectively. As shown in these figures, a flocculated structure is obvious for the deposited sediments at pH values of 2 and 4. For pH of 7, a flocculated structure is seen in which the pores between particles have been reduced. At pH values of 9 and 11 the degree of flocculation is reduced and the majority of particles are plate like for both soils (Figs 5 and 6).

Discussion

The surfaces of clay particles usually carry negative charges. When water comes in contact with clay particles, the negative electrical charges of the clay particles attract cations, including the hydrogen ions (H^+), from the surrounding water. Consequently, this leads to a thin layer of water called absorbed water that is bonded to clay particles. In addition, to the attraction of molecules of water to the surface of clay particles via exchange of ions, hydrogen bonding and Van der Waals forces are additional bonding agents that are believed to affect this. The molecules of the adsorbed water are more intense near the clay particles and the intensity of them is decreased with increasing the distance from clay particles. The combination of negatively charged clay particle surfaces and the positively charged spaces around the particles form a layer that is named diffused double layer (DDL). The adsorbed ions on the surface of clay particles may exchange places with another ion within the DDL. This transformation of ions is known as base change and the ions involved in the transformation are known as exchangeable ions (Mitchell and Soga, 2005). The nature of transformation can have a significant effect on

the properties of clay soil. For example, the thickness of DDL may be reduced as the anions exchange and cause a change in the structure of soil. Therefore, the type and amount of different cations in a clay water electrolyte system have a significant influence on the thickness of DDL.

The structure of a soil is defined as the geometric arrangement of the particles as well as the inter particle forces that may act between them. The structure of a soil has a strong influence on its mechanical behavior. The diffuse double layer can develop for individual clay particles. The DDL of neighboring clay particles interact resulting in a net repulsive force between them. Attractive forces, however, may be developed between two DDLs of particles. If the dominating forces are repulsive, it leads to dispersed structure of the soil and if they are attractive, it results in flocculated structure. Environmental effects of clay structure including pH, acidity, temperature and cations also influence the thickness of DDL.

The results indicate that the settling characteristics of the two clay soils are a function of pH of the solution. For both soils, the thickness of deposited soil in alkaline solution is more than in acidic solution. As shown in Fig.7, at pH=2 the heights of the deposited sediments are 107 and 67 mm for soils A and B, but at pH=11 they change to 240 and 134 mm respectively. It is resulted that the formed structure in both acidic or basic solutions is flocculated, but the degree of flocculation in the acidic solution is more than in the basic aqueous solution. Alumina is one of the elements that constitute the minerals of clay soil. It is amphotric and is ionized at low pH and negatively at high pH (Mitchell and Soga, 2005). As a result, interaction between the positive edge and negative surface occurs and it leads to the flocculated structure. By increasing the degree of acidity

(decreasing pH) the interaction between the edge and surface of clay plates is increased which leads to increase in the degree of flocculation of soil structure. When the aqueous solution is changed to basic, hydroxyls (OH^-) appear on the surfaces and edges of clay particles and the interaction between surface and edge of each plate of clay is reduced. This leads to a lower degree of flocculation in the clay structure in the sediments as shown in the micrographs of both soils in Figs. 5 and 6. The rate of reduction in the interaction is dependent on the pH value of the aqueous solution. A higher pH causes more reduction in the interaction of two plates of clay and degree of flocculation. These results are consistent with the findings that were reported by Kaya et al. (2006). However, they are inconsistent with the results that were reported by Van Olpen (1977) who indicated that sediment's void ratio first decreases and then increases by changing the concentration of the fluid that contains suspended particles. This trend of variations in the thickness of sediment is observed for both soils (Fig.7). As it is seen in Fig.7, at a given pH, the thickness of the deposited sediment is more for soil A than soil B. This could be due to the fact that the clay content of soil A is more than soil B as shown in Table 1. Comparing the results of variation of height of sediment with PI (plastic index that can be calculated from Atterberg limits LL and PL) shows that as PI is increased with increasing pH, the height of sediment is also increased. This can be attributed to the increasing of DDL thickness due to the repulsive forces with increasing plasticity of soil that results in increasing the inter-particle spacing.

The results in Fig.2 show that the values of liquid limit (LL) for the deposited sediments are increased with increasing pH for both soils but these variations for soil A are more than for soil B in basic aqueous conditions. It can be said in acidic condition, the degree

of flocculation is more than basic condition; therefore the particles paste to each other and form coarser particles. The specific surface of coarser particles is less than the finer particles (sediments with low degree of flocculation in basic aqueous). Therefore, the capacity of adsorbed water for sediments is decreased in acidic condition and it leads to reduction of liquid limit (see micrographs of two soils at Figs. 5 and 6).

Fig. 8 shows the compaction parameters, including maximum dry unit weight (Fig.8a) and optimum water content (Fig.8b) at the different values of pH for both soils. As shown in this figure, with increasing the pH of the aqueous solution, the optimum water content increases and the maximum dry unit weight decreases for both soils A and B. It is observed that these variations for soil A are more than soil B because of the higher clay content in soil A than soil B. The reduction in maximum dry unit weight and increase in optimum water content in the basic conditions compared with the acidic conditions can be due to the degree of flocculation and the percent of coarse particles that are formed due to the pH of the aqueous solution. In basic condition, the degree of flocculation is less (therefore pasting of particles to each other is less) than acidic condition. In other words, the percent of clay in basic condition is more than acidic condition which causes these variations in compaction parameters.

The stress-strain curves of soils A and B at different pH values are shown in Figs.4a and 4b respectively. As shown in these figures, the failure of soil A occurred at strain of about 6% but soil B failed at 3%. It is seen that the ductility of soil A is nearly twice that of soil B. This could be because the percent of clay in soil A was more than soil B (Table 1). The higher clay content of soil A increases the cementation of the soil particles which in turn prevents from the brittle behavior and increases ductility of the soil. Therefore, a

higher percent of clay increases ductility and causes the failure of the sample to occur at larger strains.

The variations of final strength of soils A and B are shown in Fig.9. It is shown that the strength of the sediments was decreased with increasing the pH of the aqueous solution for both sediments. The reduction in strength is due to the degree of flocculation of soil. For acidic condition the strength is more than for basic condition. This is because in the acidic condition the degree of flocculation is more and hence the friction between particles is more which results in greater strength than basic condition. It is seen from this figure that the final strength of the soil sediment B is less than soil A in acidic condition but for pH more than 7 (i.e pH of 9 and 11) the strength of soil B is more than soil A. It can be said that since the clay content of soil A is more than soil B, therefore the bonding between the clay particles is stronger than soil B which causes increase in its final strength. At pH values of 9 and 11, the bonding between the clay particles is not strong due to the basic condition and on the other hand for soil B the percent of silt and fine sand is more than soil A which increases the strength at these values of pH.

The results show that pH has an important effect on the behavior of sediment. Tremblay et al. (2002) indicated that the pH of pore fluid can be responsible for the cementing process and gaining strength of the sample. pH of the pore solution is important because when its value is lower than 9 hydration products are dissolved and produce no hardening or little hardening in the soil (Tremblay et al. 2002).

It was observed in this work that the behaviors of the two clay soils at the same pH are not same. Also, the behavior of each of the soil is different at the different pH values. It can be said that the behavior of a deposited soil is dependent on the type of clay minerals,

percent of clay, specific surface of clay particles and chemical quality of the fluid in which the deposition has occurred. Therefore, the results obtained for a particular soil cannot be generalized to other soils. Conducting appropriate experimental tests is an essential step for understanding the characteristics of a particular deposited sediment.

Sediments may be formed by seasonal rivers or lakes or from dredged materials from harbors or waterways that are disposed in land. Dubois et al. (2009) reported that about 100 million tones of materials are dredged around the world and disposed in land. They have specific characteristics that differ from region to region. Deposition of large amounts of these materials may cause new environmental problems. However, they can be considered as a new source of materials and can be used in civil engineering applications. The laboratory characterization of these materials is an essential step in their management. Physical, mechanical, mineralogical and environmental properties of sediments define their possible usage in civil engineering. Today they are used as road construction materials (Wang et al. 2017), clay bricks (Baksa et al., 2018), cementitious materials and paving blocks (Dubois et al. 2009).

Conclusion

A set of experimental tests were conducted on two different soils that were settled in aqueous solutions with different pH values. Based on the results, the following conclusions can be drawn:

- The engineering properties of sediments such as Atterberg limits, compaction parameters and strength are dependent on the nature of the formed sediments.

- As the pH of the aqueous solution becomes basic the final sediment thickness is increased but for acidic conditions it is decreased.

- The Atterberg limits and compaction parameters (maximum dry unit weight and optimum water content) for both soils are dependent on the pH of the aqueous solution in which the settlement takes place. In addition, the type of soil minerals influences the variations of these parameters.
- The strength of sediments that were formed in acidic aqueous solutions is increased in comparison with the sediments formed in basic aqueous solutions.

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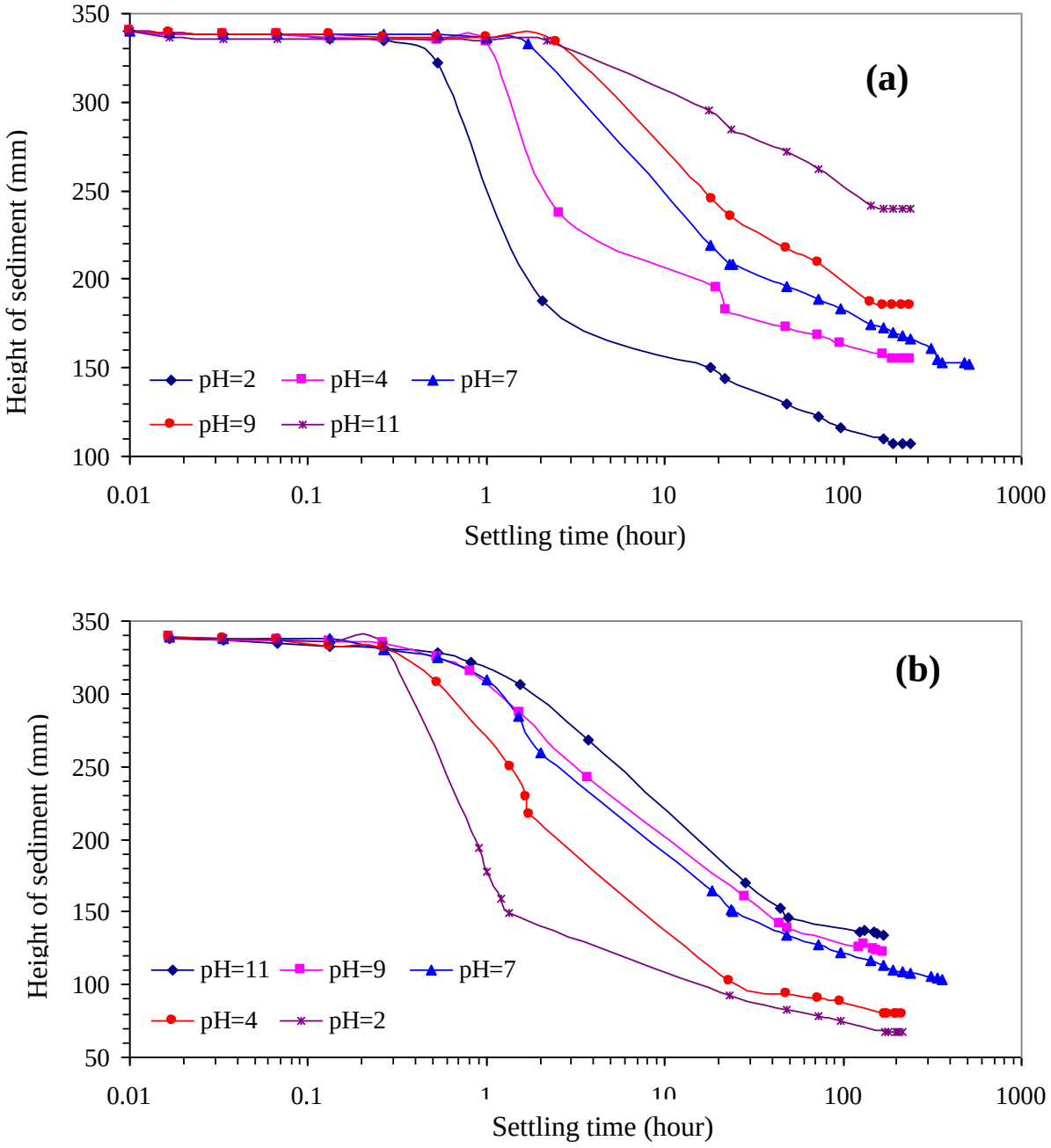


Fig.1. Settling behavior of (a) soil A and (b) soil B with time at different pH values

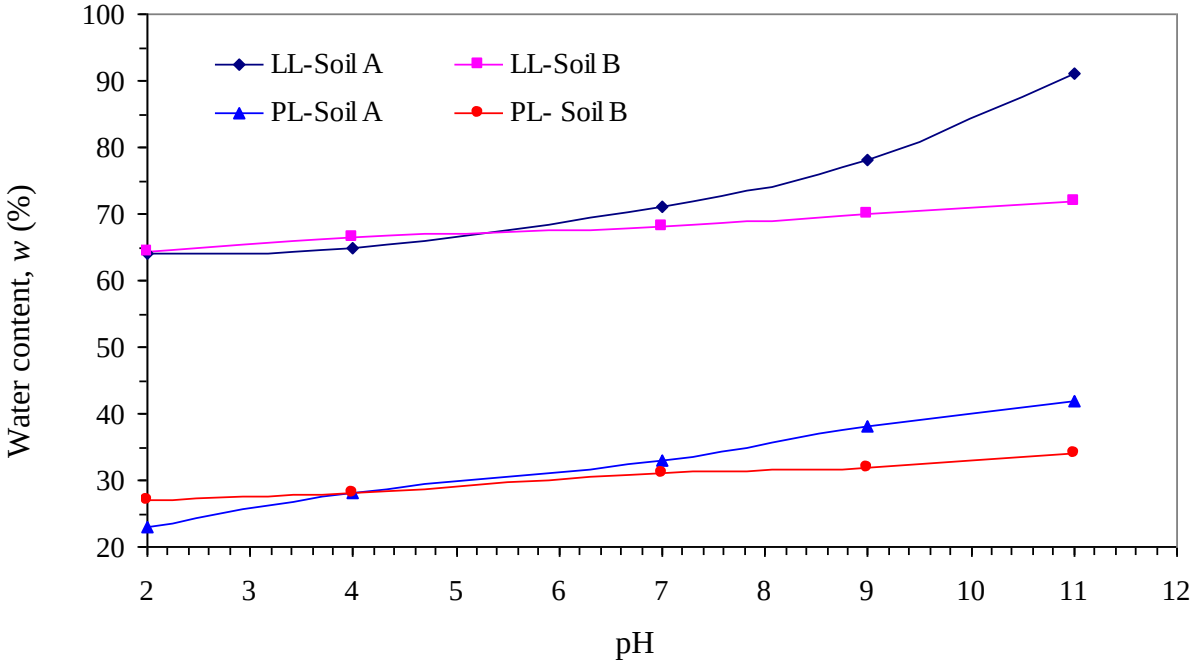
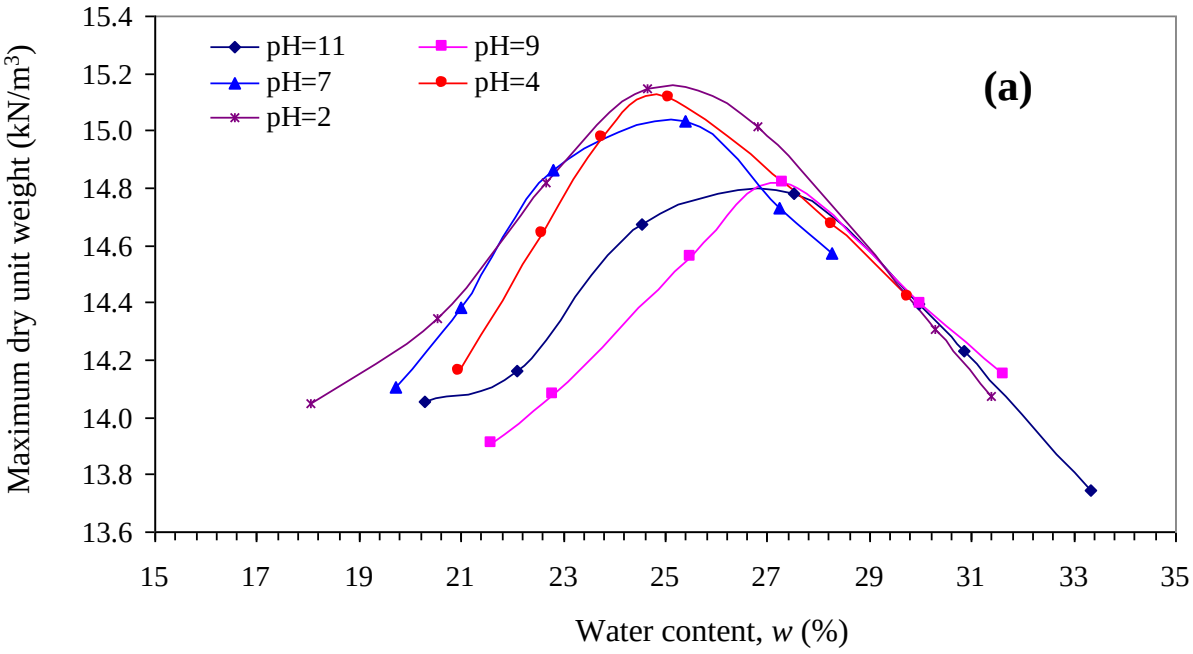


Fig.2. Variations of liquid limit (LL) and plastic limit (PL) for soils A and B at various pH values



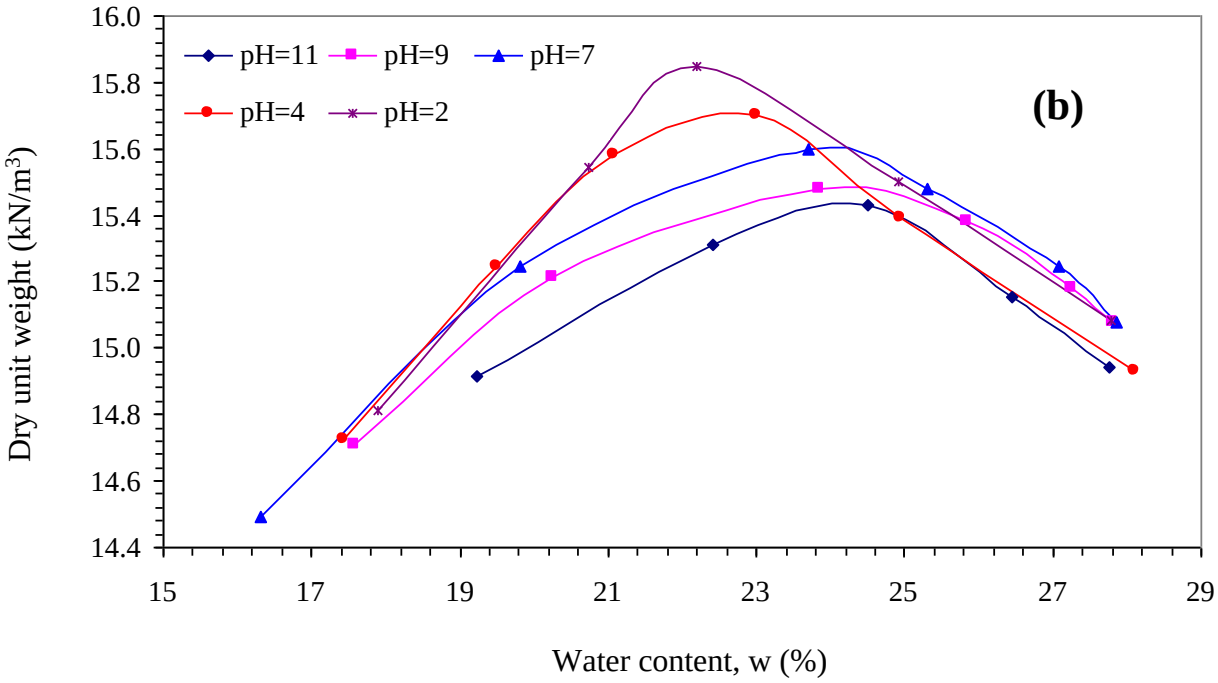
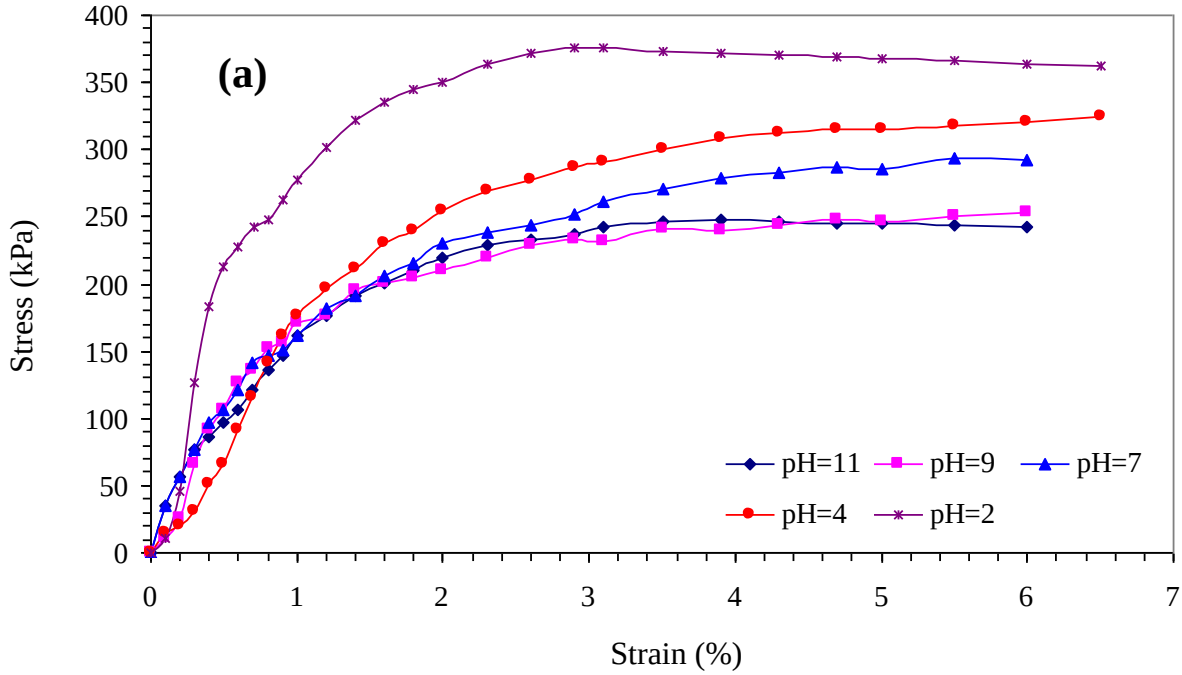


Fig.3. Compaction curves for soil A (a) and Soil B (b) at different pH values



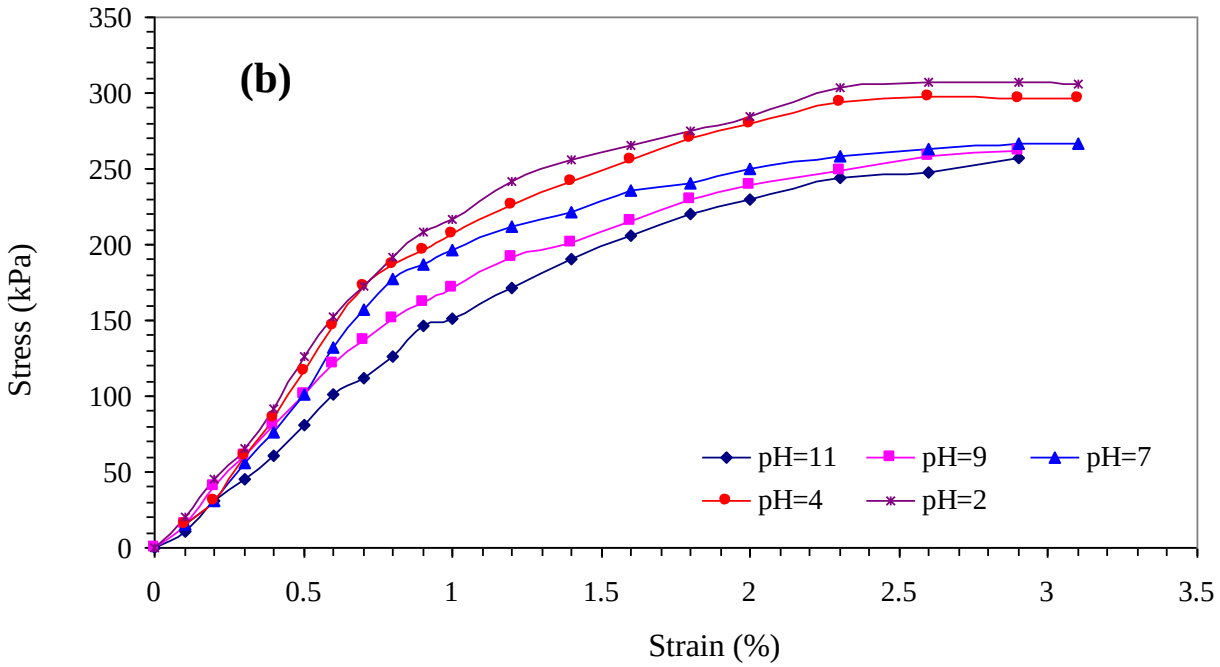


Fig.4. Stress-strain curves for (a) soil A and (b) soil B for different values of pH

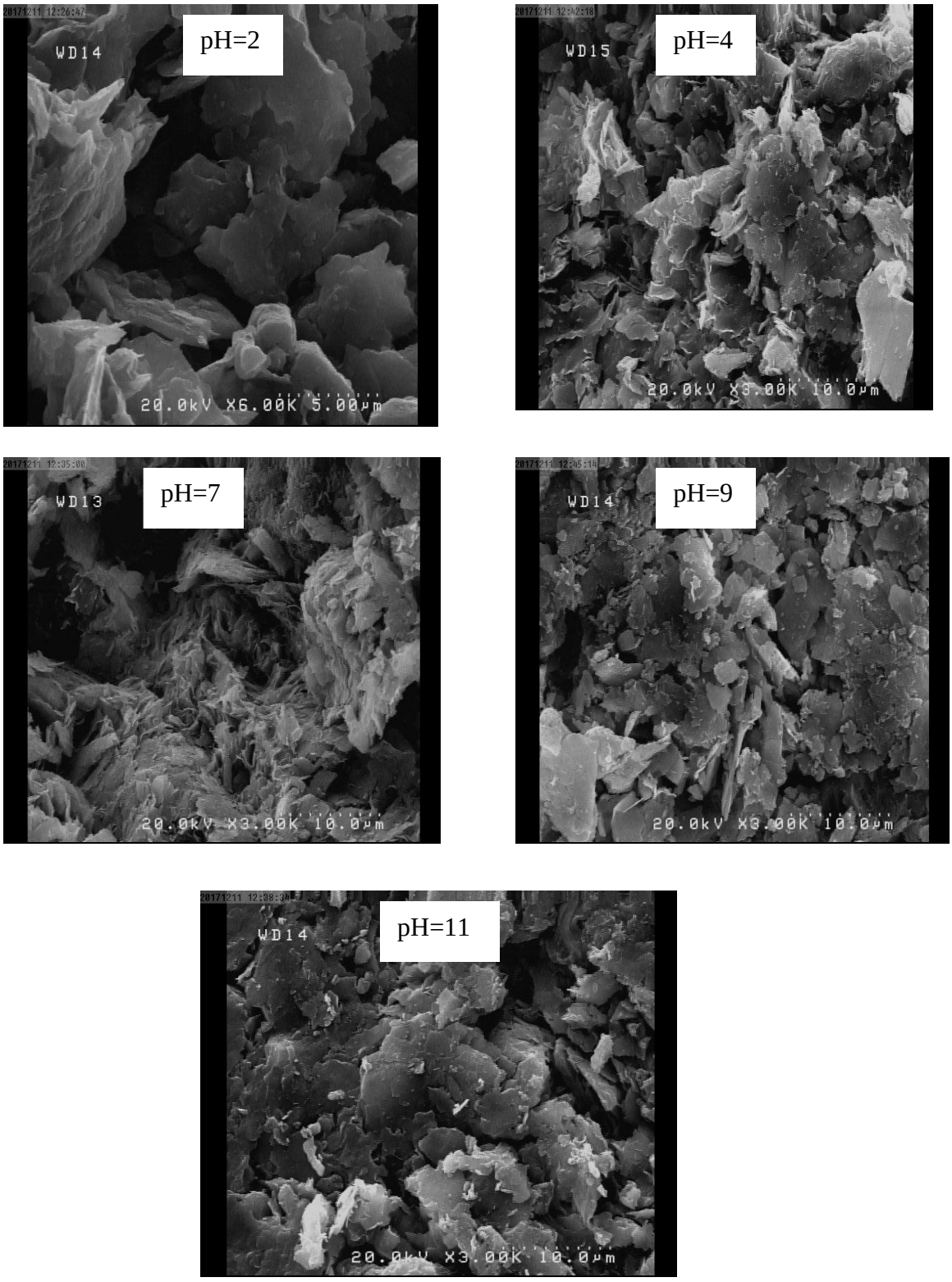


Fig.5. Scanning electron micrograph of soil A at different values of pH

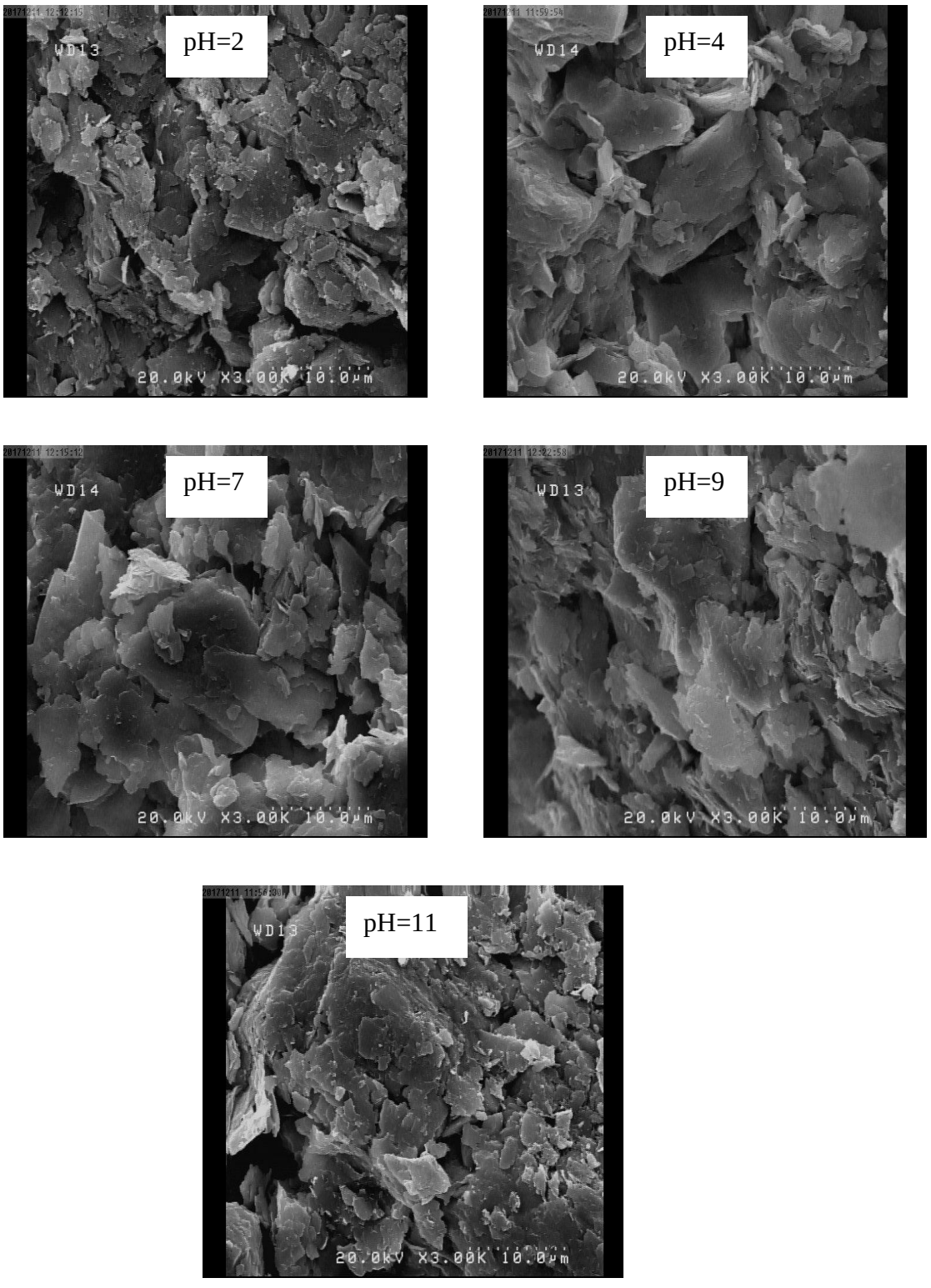


Fig.6. Scanning electron micrograph for soil B at different values of pH

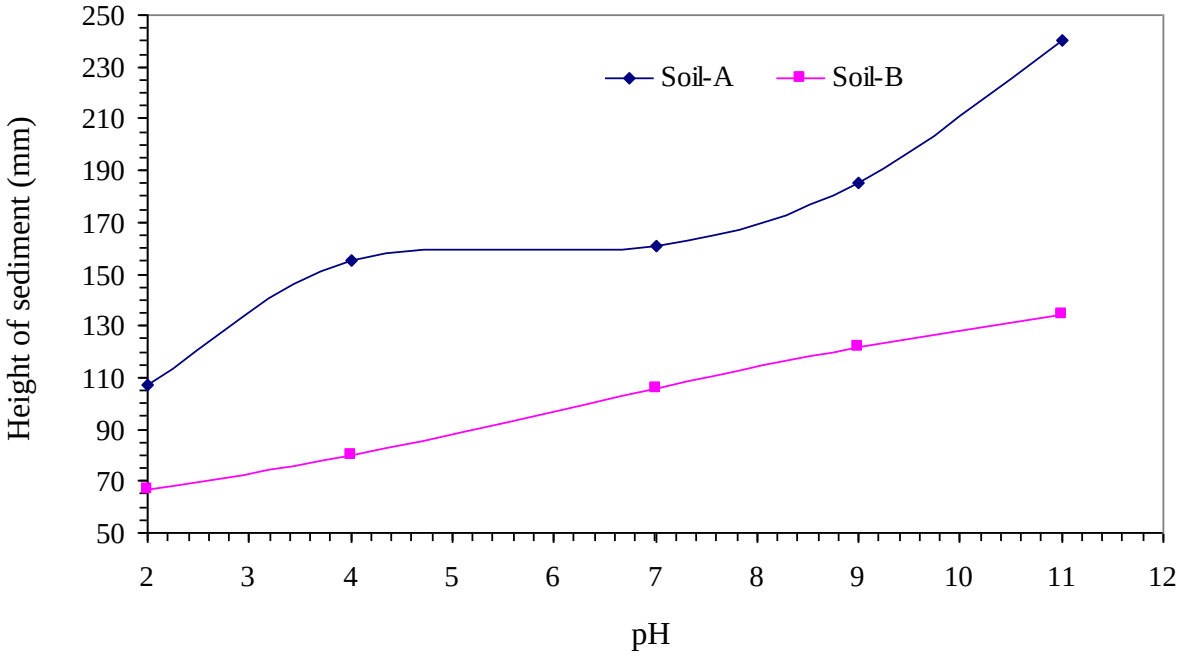
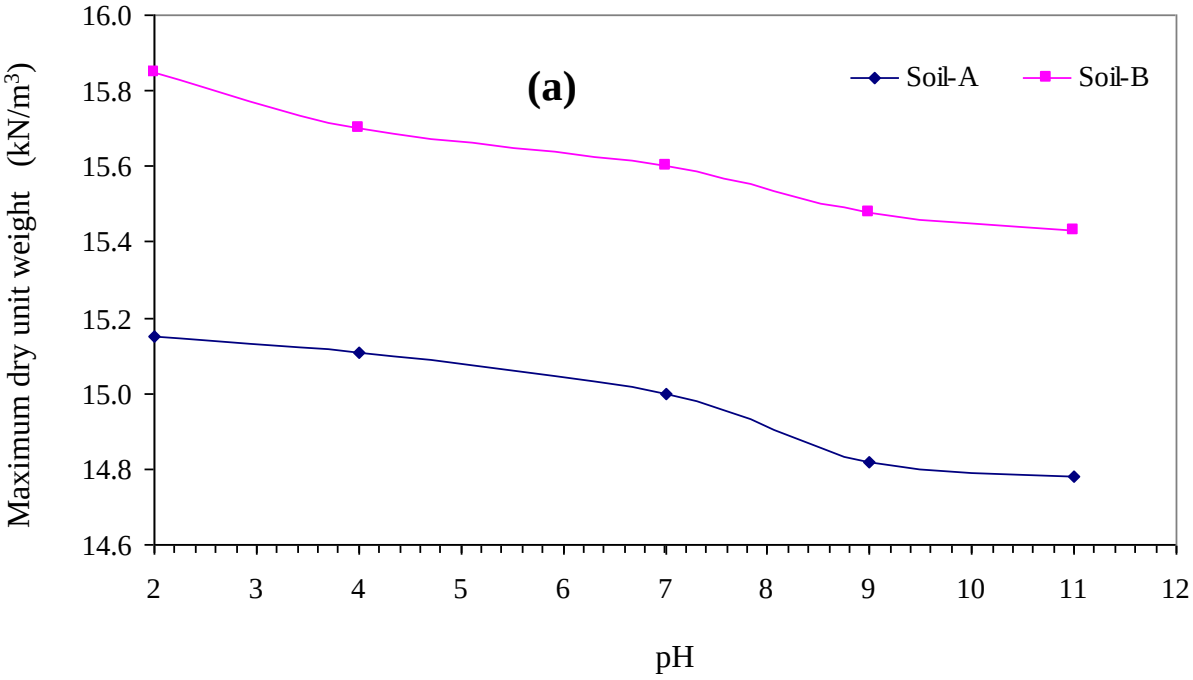


Fig.7. Final height of sediment at different pH values for soil A and soil B



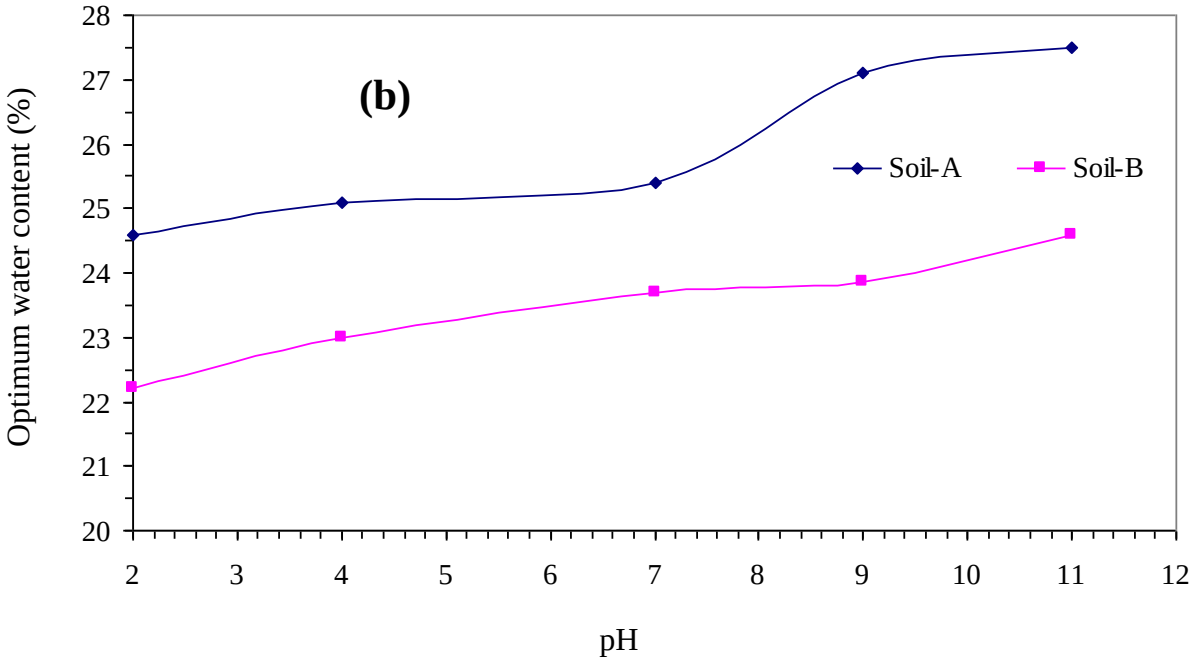


Fig.8. Variations of (a) maximum dry unit weight and (b) optimum water content against pH for sediments of soils A and B

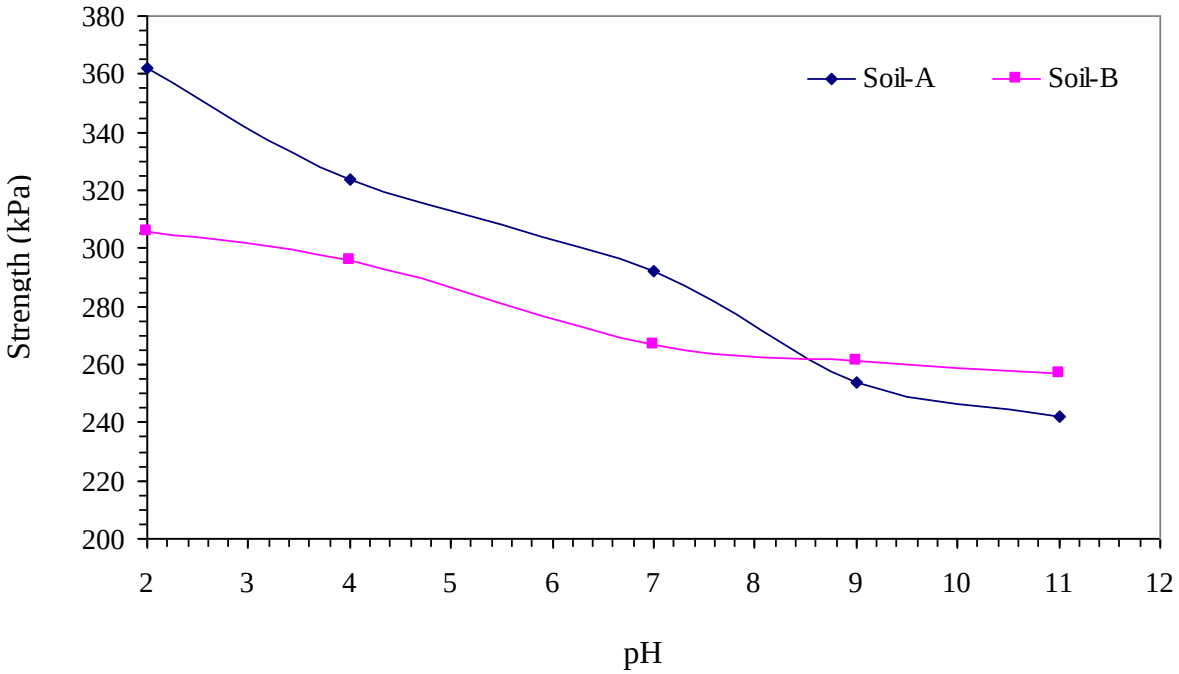


Fig.9. Final strength of sediment of soils A and B at different pH values

Table 1. Physical and mechanical properties of soils

Properties	Soil A	Soil B
G_s	2.75	2.73
Consistency limits		
LL (%)	83	70
PL (%)	35	25
PI (%)	48	45
Particle distribution		
Sand (%)	0.0	0.0
Silt (%)	80	88
Clay (%)	20	12
USCS classification	CH	CH
Compaction characteristics		
Optimum water content, *w* (%)	28.8	25
Maximum dry unit weight, γ_{dmax} (kN/m^3)	14.4	15.7